LLNL-JRNL-561992
LLNL-JRNL-574772

Corrosion-resistant, triple-wavelength Mg/SiC multilayer coatings for the 25-80 nm wavelength region

Regina Soufli
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2012 International Workshop on EUV and Soft X-ray Sources, University College Dublin, Ireland
October 9, 2012
This work was performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344.

Contributors

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 Luis Rodríguez-De Marcos, Jose A. Méndez, Juan I. Larruquert

- ***Lawrence Berkeley National Lab*:** Eric M. Gullikson

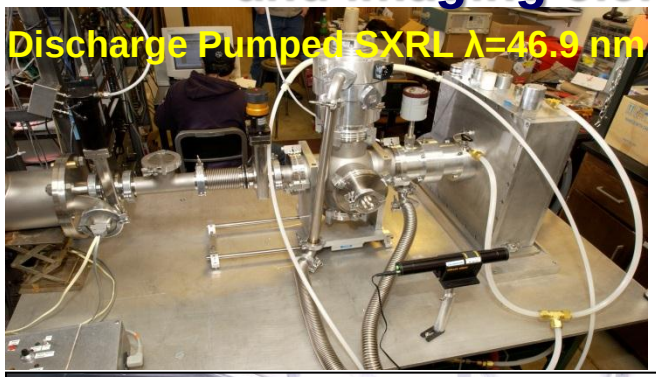
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Overview

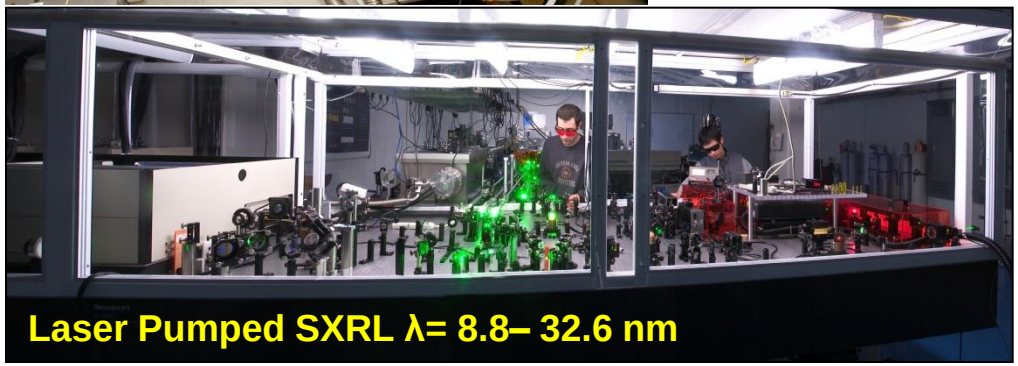
- Motivation: applications that could benefit from Mg/SiC multilayers with high performance and long lifetime
- Design and experimental performance of triple-wavelength Mg/SiC multilayers
- Origins, propagation mechanisms and impact of Mg/SiC corrosion
- Corrosion barriers for Mg/SiC multilayers
- Design and experimental performance of triple-wavelength Mg/SiC multilayers with corrosion barriers
- Summary and future work

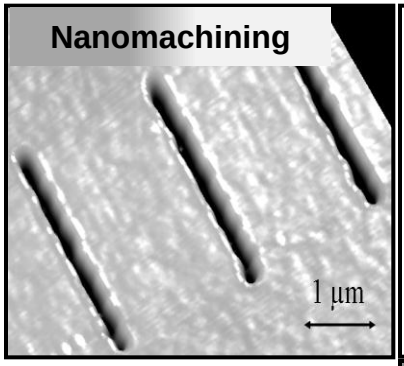
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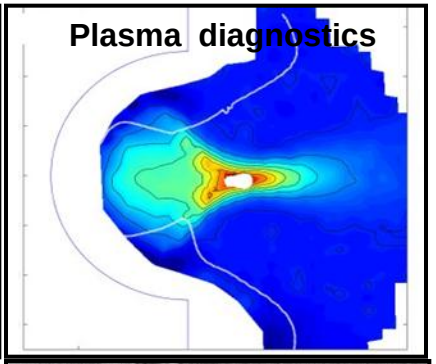
Compact plasma-based EUV/soft x-ray laser applications need multilayer coatings as collector and imaging elements

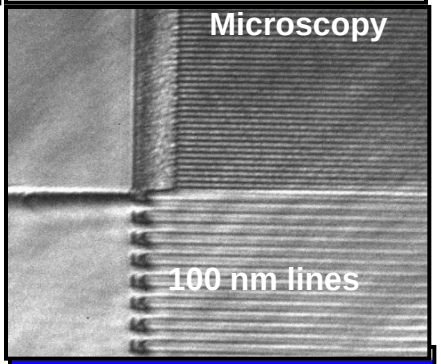


Courtesy: Jorge Rocca and Carmen Menoni, Colorado State University

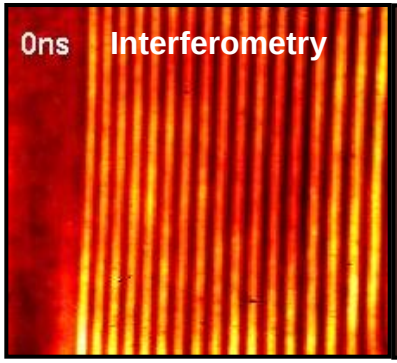


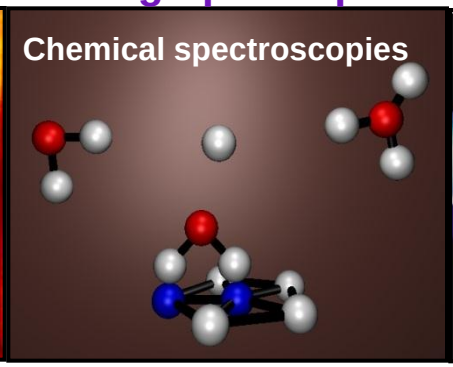


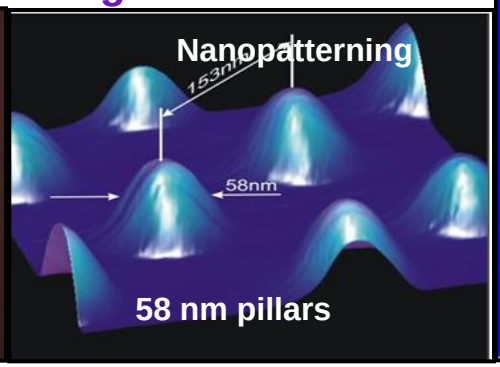


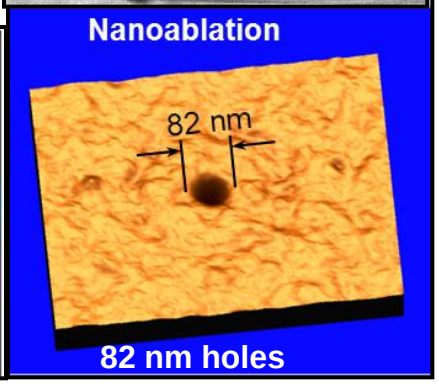


- High pulse energy (μJ-mJ)
- High monochromaticity ($\lambda/\Delta\lambda < 10^{-4}$)
- High peak spectral brightness

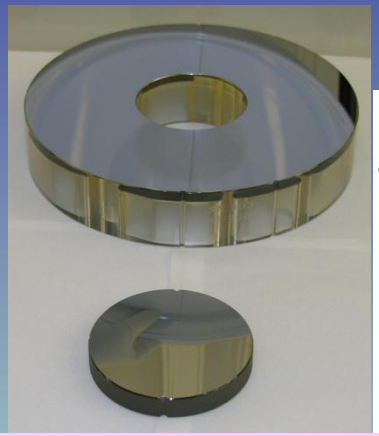




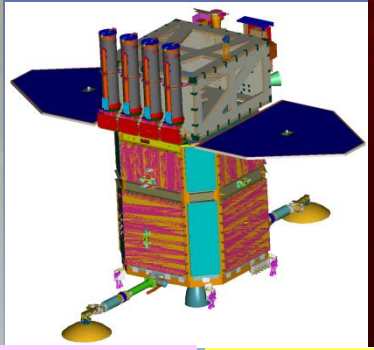


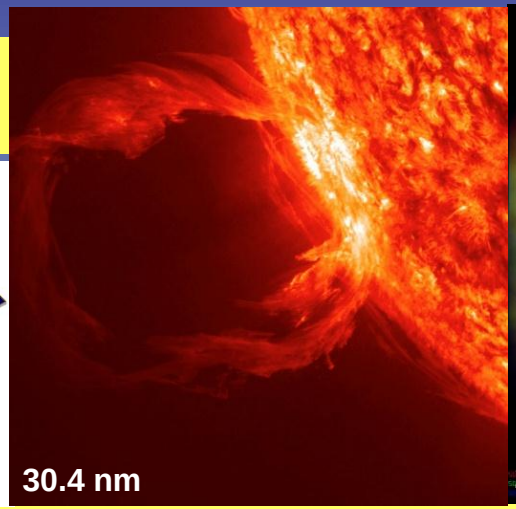


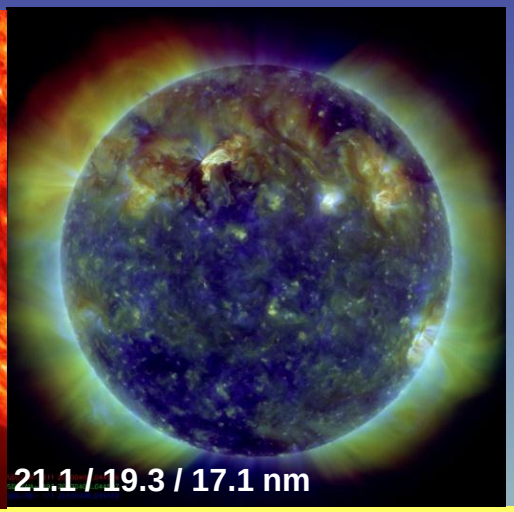
Multilayers with high performance and long lifetime are needed for EUV solar physics and space weather missions



7 EUV wavelengths,
9.4 nm to 33.5 nm.
Mo/Y, Mo/Si, SiC/Si.

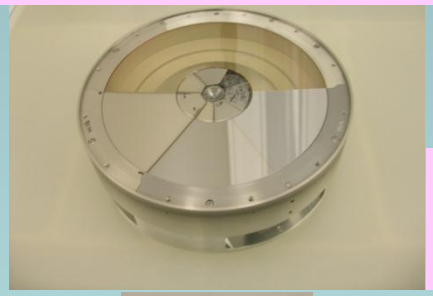


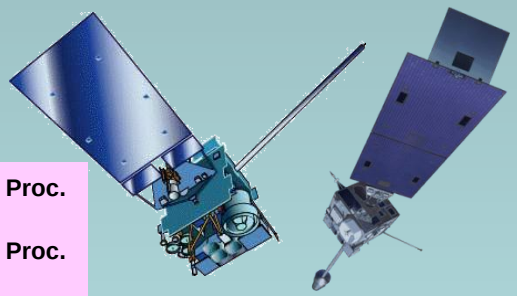




R. Soufli, *et al.*, Proc. SPIE 5901, 59010M (2005)
R. Soufli, *et al.*, Appl. Opt. 46, 3156-3163 (2007)
P. Boerner *et al*, Solar Physics 275, 41-66 (2012)
J. R. Lemen *et al*, Solar Physics 275, 17-40 (2012)
R. Soufli, *et al.*, Proc. SPIE 8443, 8443R (2012)

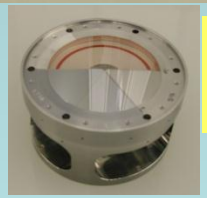
NASA's Solar Dynamics Observatory (SDO). Launch date: February 11, 2010.
http://sdo.gsfc.nasa.gov







D. Martinez-Galarce, *et al*, Proc. SPIE 7732, 773237 (2010).
D. Martinez-Galarce, *et al*, Proc. SPIE 8501 (2012).

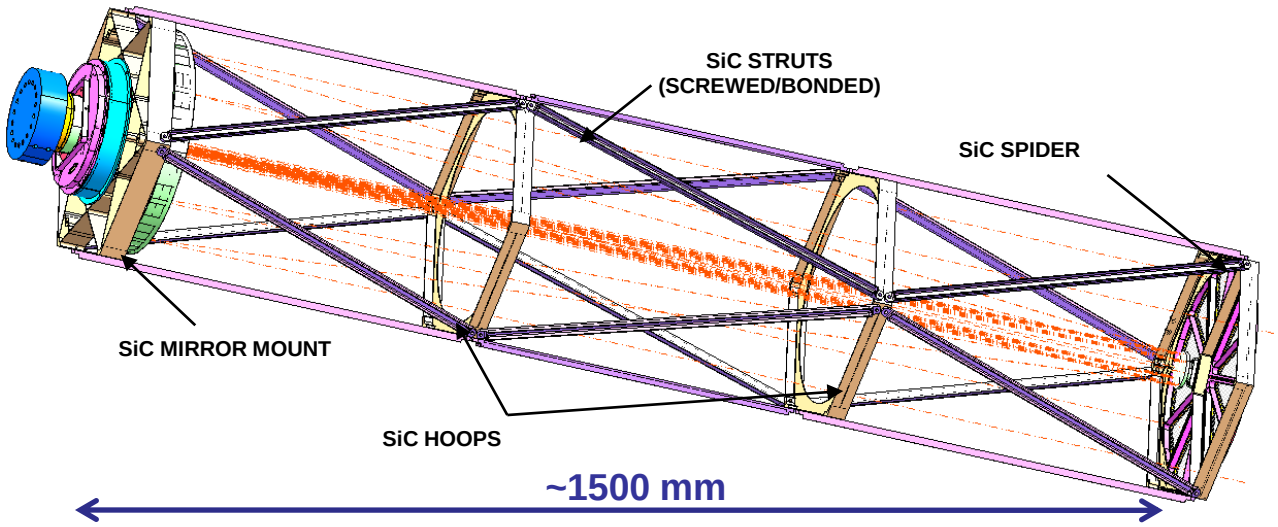


Multilayer-coated test mirrors for NASA/NOAA's GOES-R space weather satellite. 6 EUV wavelengths, 9.4 nm to 30.4 nm (Mo/Y, Mo/Si).
Launch date: 2014

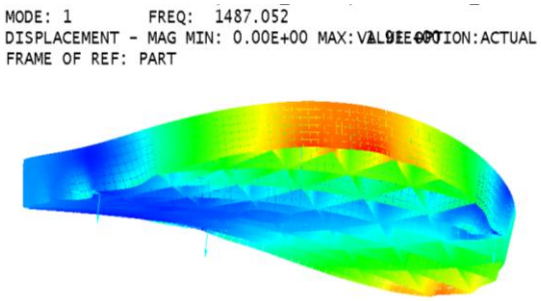
Reflective X-rayOptics

LOCKHEED MARTIN

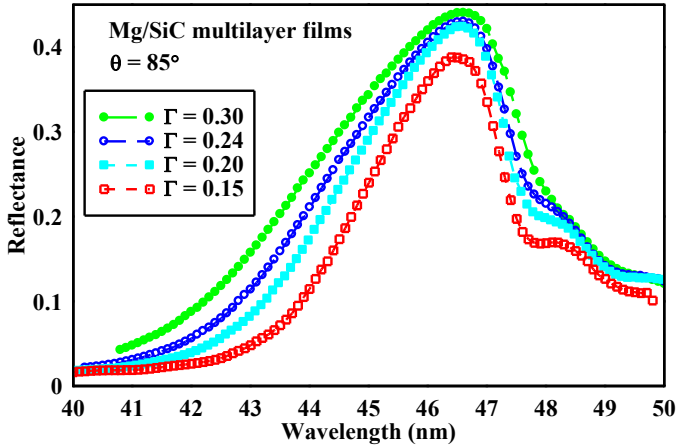
Multilayer coatings are needed for future solar missions designed to operate at 46.5 nm



HiLiTE: a 300-mm aperture Cassegrain telescope, aiming to study the Sun's transition region at the 46.5 nm Ne VII emission line. Designed to be made entirely of SiC, including optical substrates and metering structure.



	System
Effective Focal Length	10.313 m
Focal Ratio	f/34
Plate Scale	20 arcsec/mm
Field of View	> 4x4 arcmin*
	Primary Mirror
Clear Aperture	300 mm
Radius of Curvature	- 3564.2564 mm
Conic	- 1
	Secondary Mirror
Clear Aperture	40 mm
Radius of Curvature	- 680.89795 mm
Conic	- 2.0116



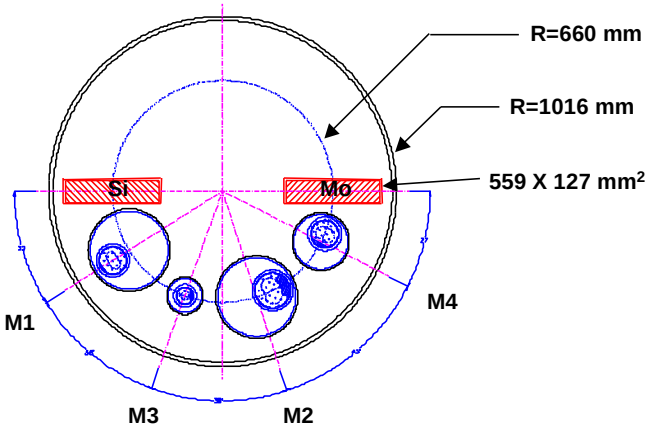
D. S. Martínez-Galarce, P. Boerner, R. Soufli, B. De Pontieu, N. Katz, A. Title, E. M. Gullikson, J. C. Robinson, S. L. Baker, "The high-resolution lightweight telescope for the EUV (HiLiTE)", Proc. SPIE 7011 , 70113K (2008).

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LLNL facilities used for multilayer deposition and characterization of Mg/SiC multilayers discussed in this presentation

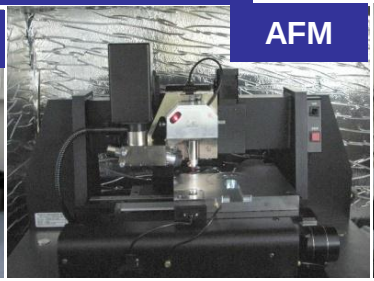
DC- magnetron sputtering multilayer deposition system





Precision surface metrology





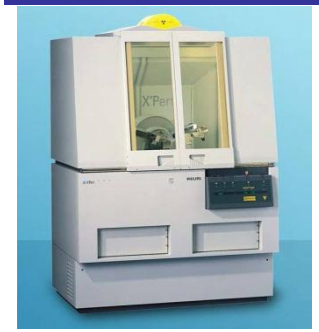


Also (not pictured):
- Contact profilometers
- Thin film stress measurement apparatus

Custom cleaning facility for optical substrates



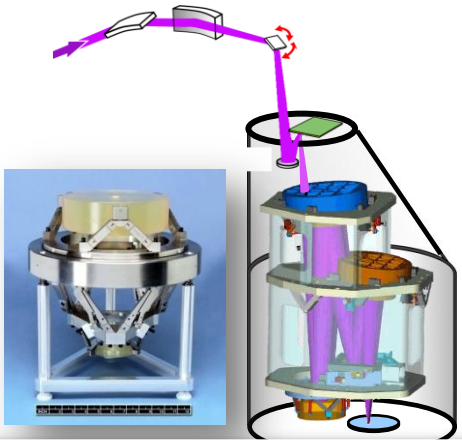
X-Ray Diffractometer



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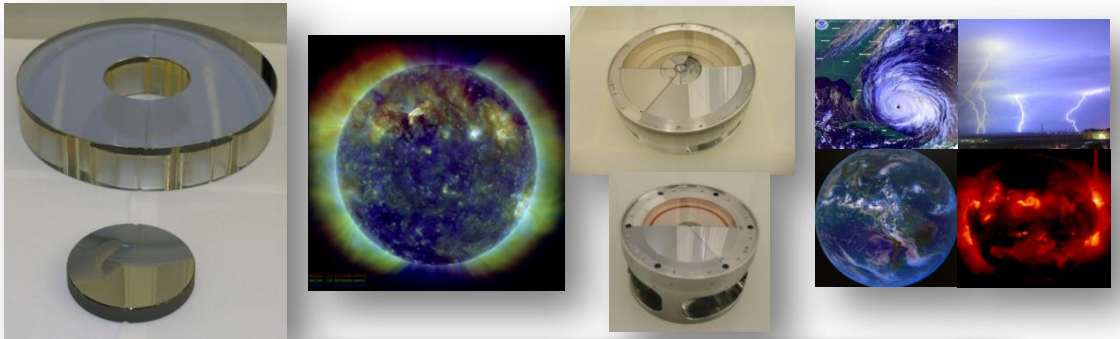
Our group at LLNL has participated in the development of optics for a wide range of applications for EUV, soft and hard x-ray wavelengths

EUV Lithography



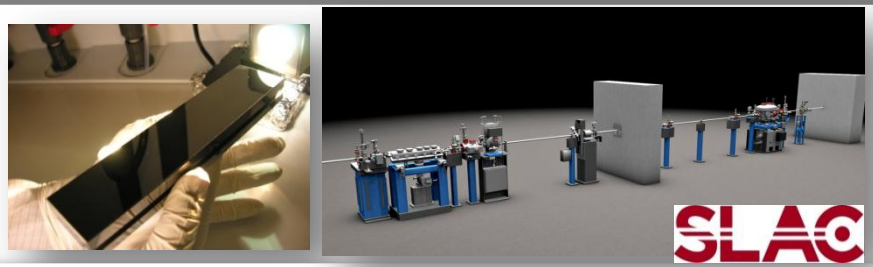
R. Soufli *et al*., *Proc. SPIE* 4343, 51 (2001)
R. Soufli *et al*., *Appl. Opt.* 46, 3736 (2007)

EUV solar missions (NASA's SDO and NASA/NOAA's GOES-R)



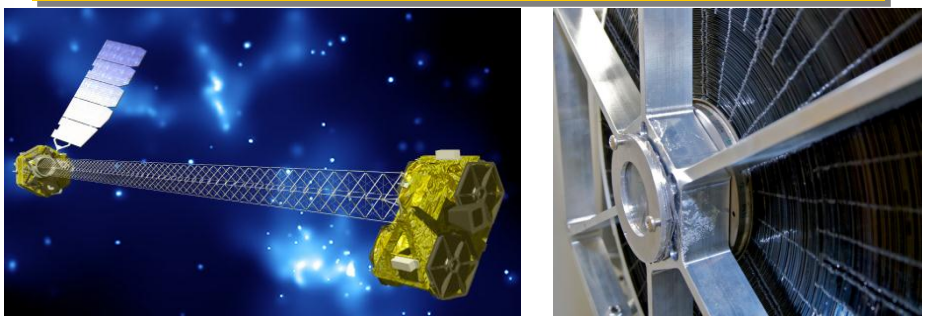
R. Soufli, *et al*., Proc. SPIE 5901, 59010M (2005)
R. Soufli, *et al*., Appl. Opt. 46, 3156-3163 (2007)
P. Boerner *et al*, Solar Physics 275, 41-66 (2012)
J. R. Lemen *et al*, Solar Physics 275, 17-40 (2012)
R. Soufli, *et al*., Proc. SPIE 8443 8443R (2012)

X-ray optics for the LCLS free-electron laser



R. Soufli, M. Fernandez-Perea *et al*., Appl. Opt. 51, 2118 (2012)
R. Soufli, M. Fernandez-Perea *et al*., Proc. SPIE 8077, 807702 (2011)
R. Soufli *et al*., Proc. SPIE 7361, 73610U (2009)
A. Barty, R. Soufli *et al*., Optics Express 17, 15508-15519 (2009)
R. Soufli, A. L. Aquila *et al*., Appl. Opt. 47, 4633-4639 (2008)
R. Soufli, M. J. Pivovaroff *et al*., Proc. SPIE 7077, 707716 (2008)
T. J. McCarville *et al*., Proc. SPIE 7077, 7077E (2008)
M. J. Pivovaroff *et al*., Proc. SPIE 6705, 6705O (2007)

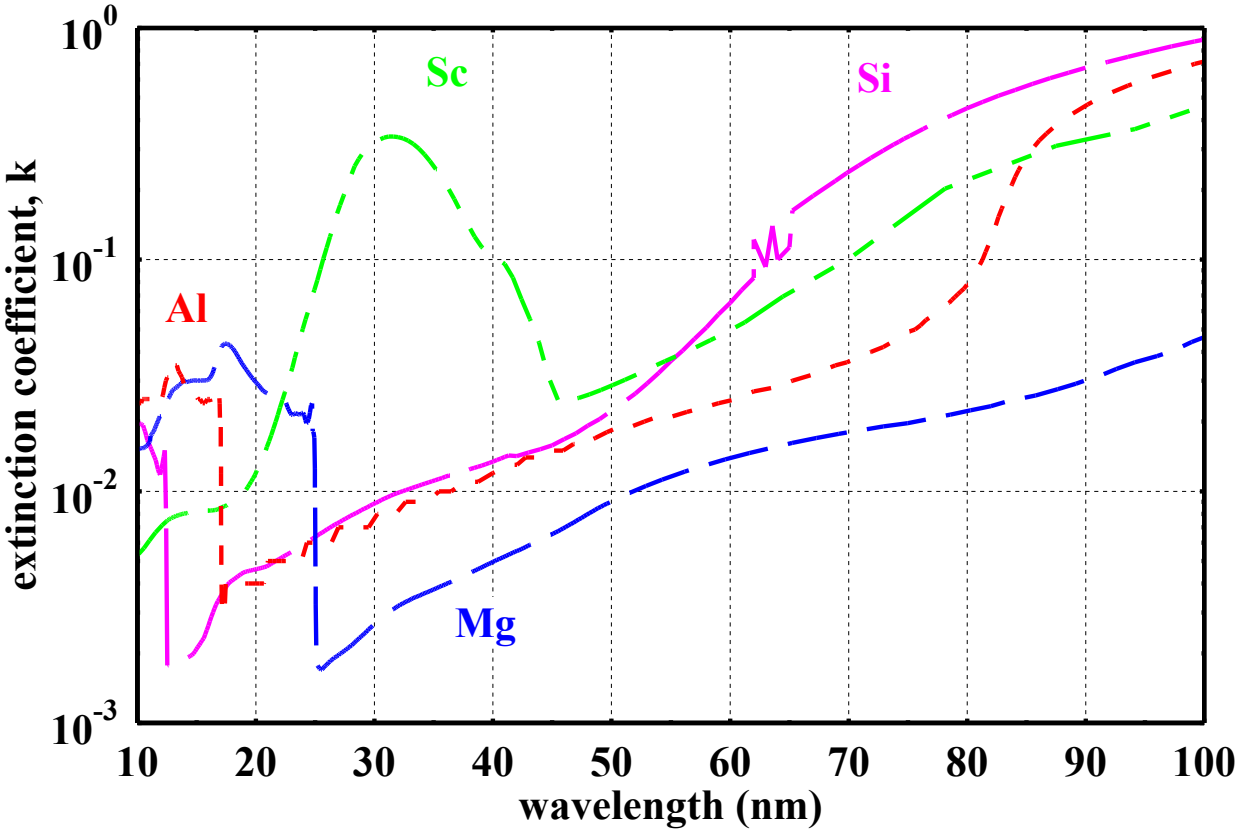
Hard x-ray space missions (NASA's NuSTAR



C. J. Hailey *et al*. Proc. SPIE 7732, 77320T (2010)
F. A. Harrison *et al*. Proc. SPIE 7732, 77320S (2010)

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Mg exhibits consistently low absorption in an extended wavelength range, longer than the Mg $L_{2,3}$ edge (25 nm)



Refractive index = *n* + *i*k*

Extinction coefficient *k* values obtained from:

1. Mg: M. Vidal-Dasilva, *et al*, J. Appl. Phys. 108, 063517 (2010).

2. Al: E. Shiles, *et al,* Phys. Rev. B 22, 1612-1628 (1980), as compiled by E. D. Palik, *Handbook of optical constants of solids* (Academic Press, 1985).

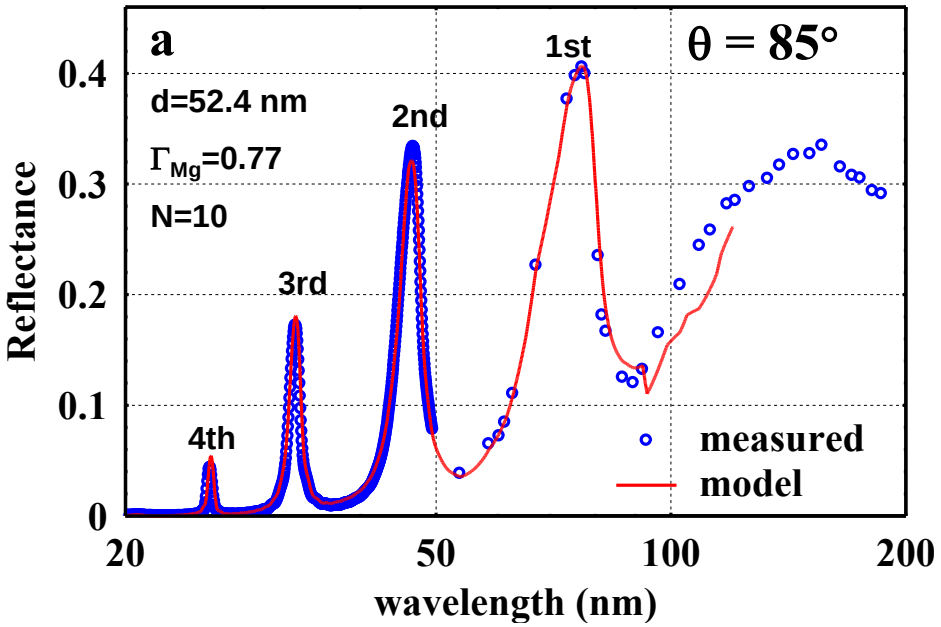
3. Si: R. Soufli and E. M. Gullikson, Appl. Opt. 36, 5499-5507 (1997).
H. R. Philipp, J. Appl. Phys. 43, 2835-2839 (1972), as compiled by E. D. Palik, *Handbook of optical constants of solids* (Academic Press, 1985).

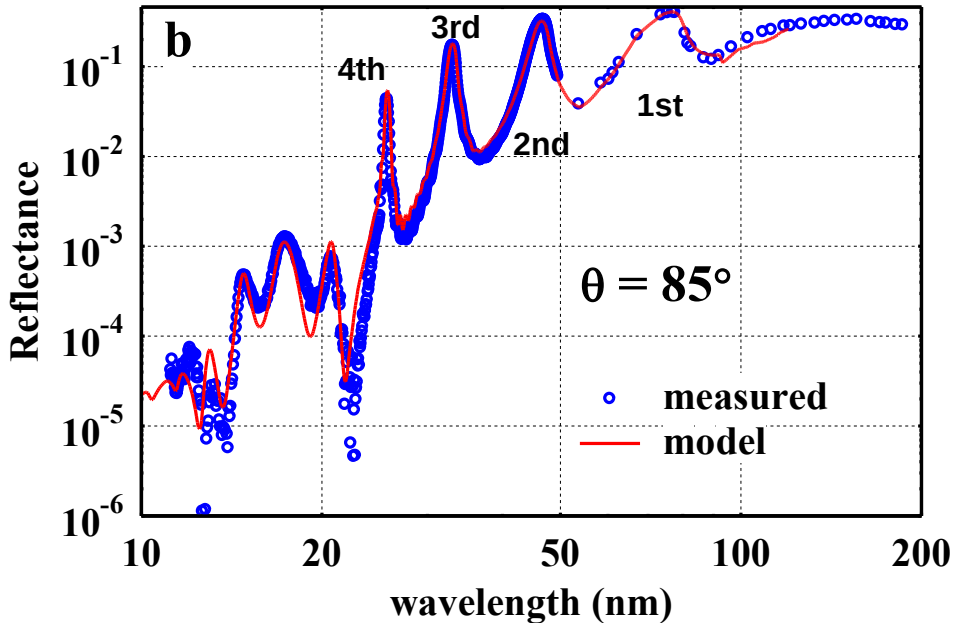
4. Sc: M. Fernández-Perea, *et al,* J. Opt. Soc. Am. A 23, 2880-2887 (2006).

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We have achieved the highest reported narrowband peak reflectance 40.6% at 76.9 nm and near-normal incidence

Standard Mg/SiC (no corrosion barriers)





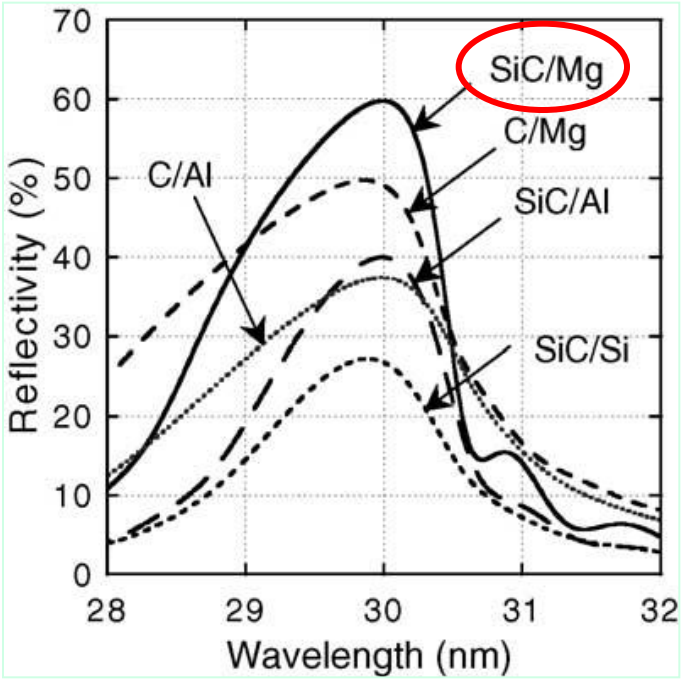
- ❑ Mesurements performed at:
- Beamline 6.3.2., Advanced Light Source, LBNL, for < 50 nm wavelengths
- GOLD facility, Instituto de Óptica, Madrid, Spain, for 50-200 nm wavelengths
- ❑ Model = IMD software by D. L. Windt, Computers in Physics 12, 360–370 (1998)

M. Fernández-Perea, R. Soufli, J. C. Robinson, L. Rodríguez-de Marcos, J. A. Mendez, J. I. Larruquert and E. M. Gullikson, "Triple-wavelength, narrowband Mg/SiC multilayers with corrosion barriers and high peak reflectance in the 25-80 nm wavelength region", Optics Express **20,** 24018-24029 (2012).

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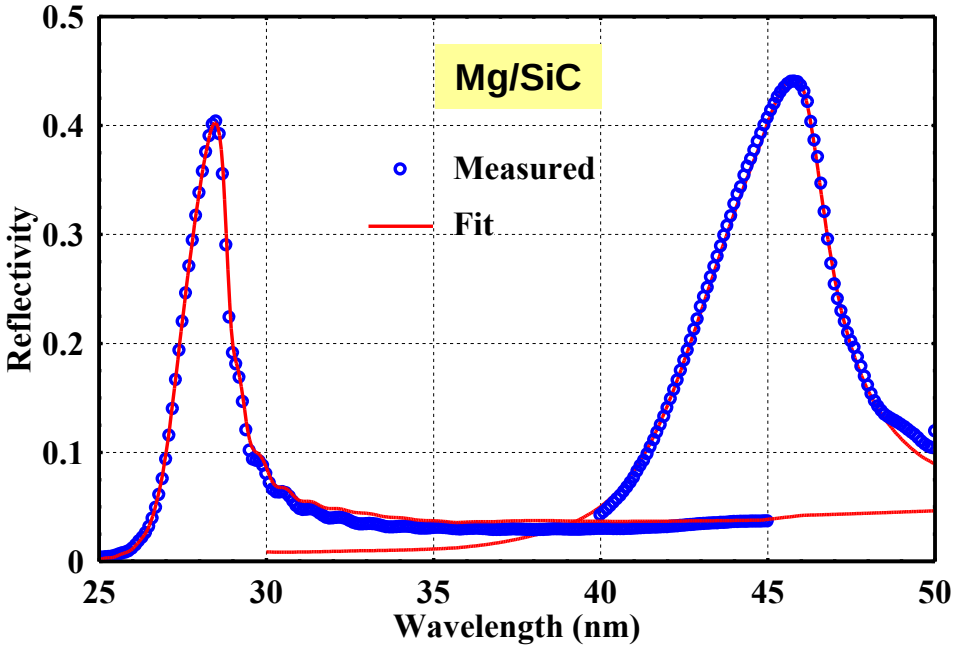
Mg/SiC is the best performing multilayer in the 25-80 nm wavelength region, except ….

Calculated ideal reflectivity (roughness=0) for different multilayers



H. Takenaka *et al.*, J.of El. Spectr. and Rel. Phen. **144-147**, 1047 (2005).

Experimental demonstration and modeling of SiC/Mg multilayers deposited at LLNL



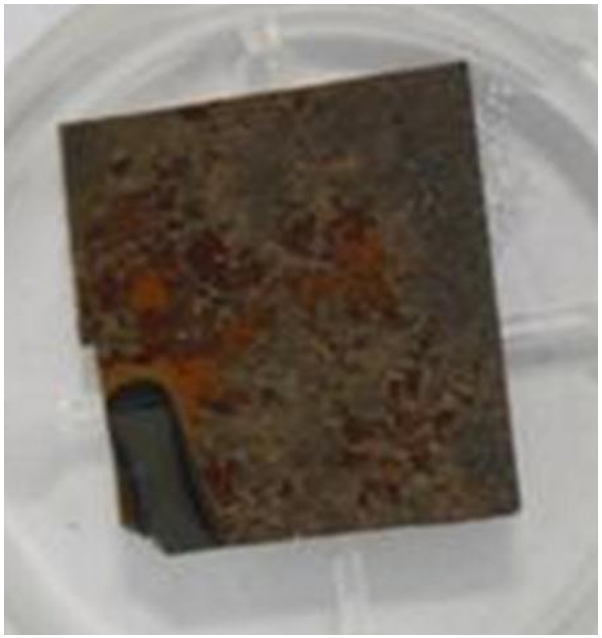
Measured at ALS beamline 6.3.2 (LBNL)

Mg/SiC exhibits a unique combination of high reflectivity, near-zero stress, thermal stability to ~350° C and good spectral selectivity compared to other candidate multilayer pairs in the 25-80 nm region

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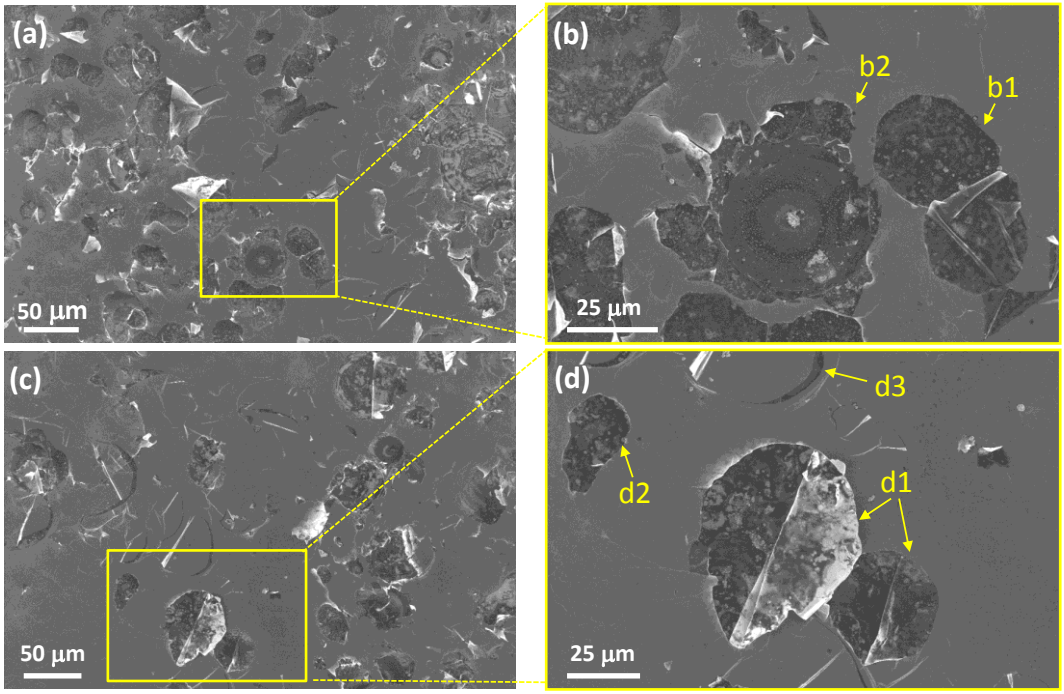
…atmospheric corrosion prevents Mg/SiC from being used in applications requiring long lifetime stability

Mg/SiC with advanced corrosion aged for 4 years



M. G. Pelizzo, A. J. Corso, P. Zuppella, P. Nicolosi, S. Fineschi, J. Seely, B. Kjornrattanawanich, and D. L. Windt, "Long-term stability of Mg/SiC multilayers", *Opt. Eng.* **51**, 023801 (2012).

Top-surface SEM of Mg/SiC with advanced corrosion aged for 2.7 years

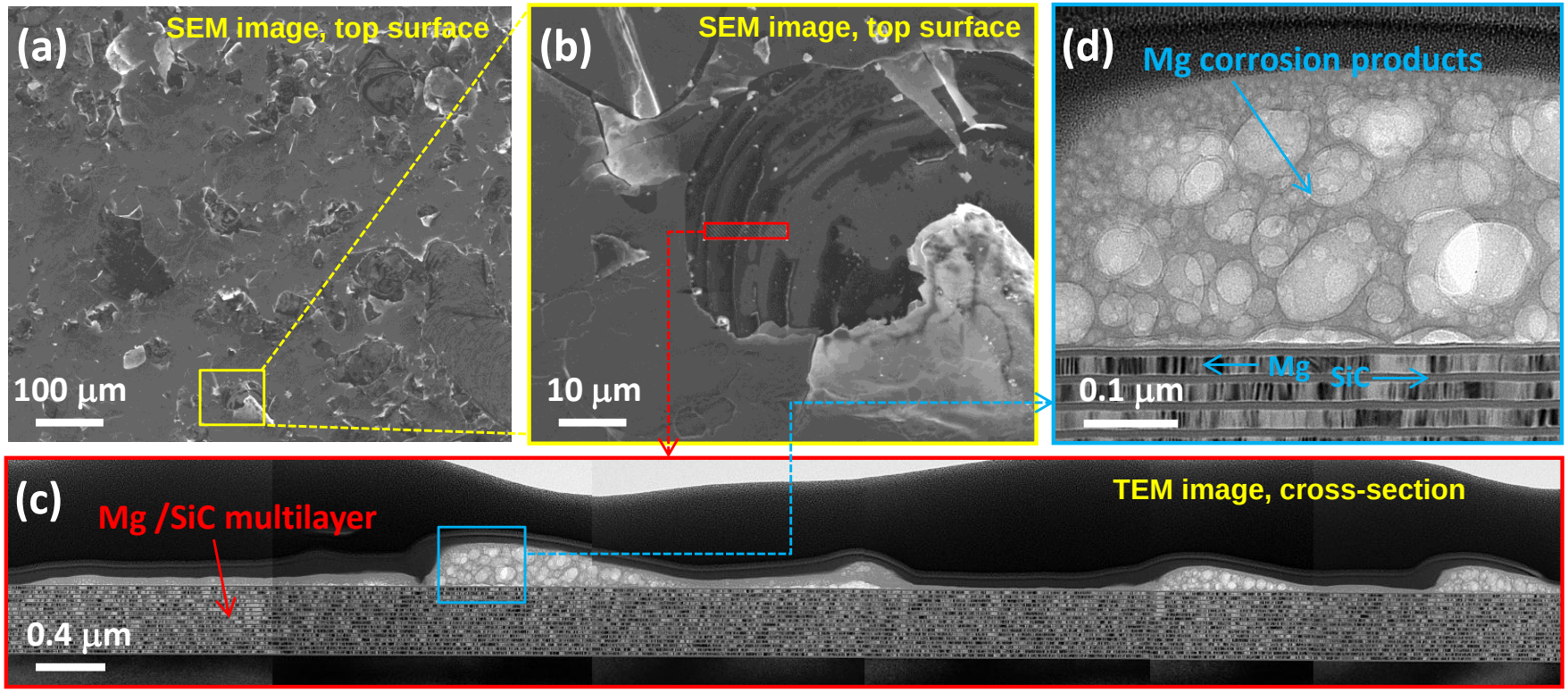


R. Soufli, M. Fernández-Perea, S. L. Baker, J. C. Robinson, J. Alameda, C. C. Walton, 'Spontaneously intermixed Al-Mg barriers enable corrosion-resistant Mg/SiC multilayer coatings', App. Phys. Lett. **101**, 043111 (2012).

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Advanced corrosion exhibits eruptive effects due to formation and volume expansion of corrosion products

Mg/SiC with advanced, eruptive stages of corrosion aged for 3 years.
4 top bilayers (out of 20) have been consumed and are missing



Binding energy (eV)	Orbital	Species
50.8	Mg 2p	MgO, $Mg(OH)_2$, $Mg(CO)_3$
532.8	O 1s	OH^-, $Mg(OH)_2$

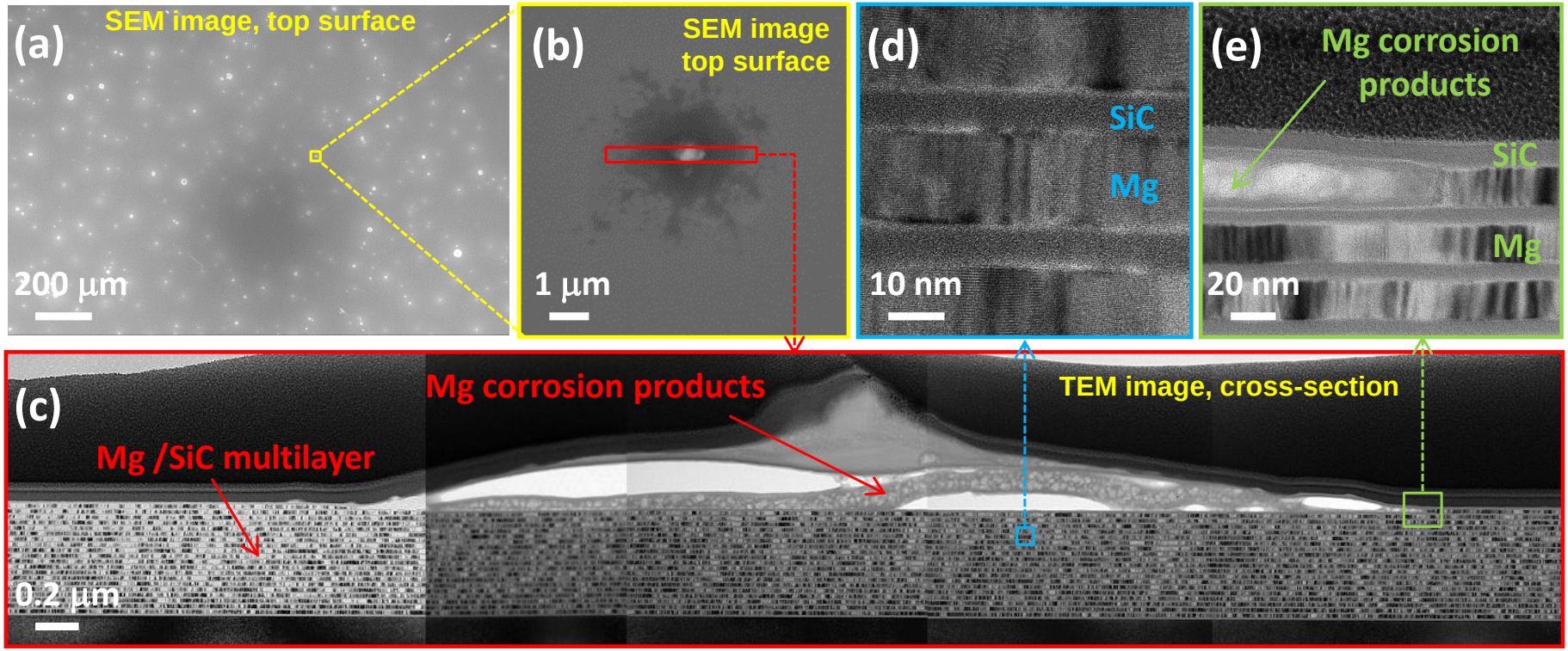
TEM and XPS analysis performed at EAG Labs, Sunnyvale, California

9 October 2012

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We have elucidated the origins and propagation mechanisms of corrosion in Mg/SiC multilayers

Mg/SiC aged for 3 years, with early, pre-eruptive stages of corrosion

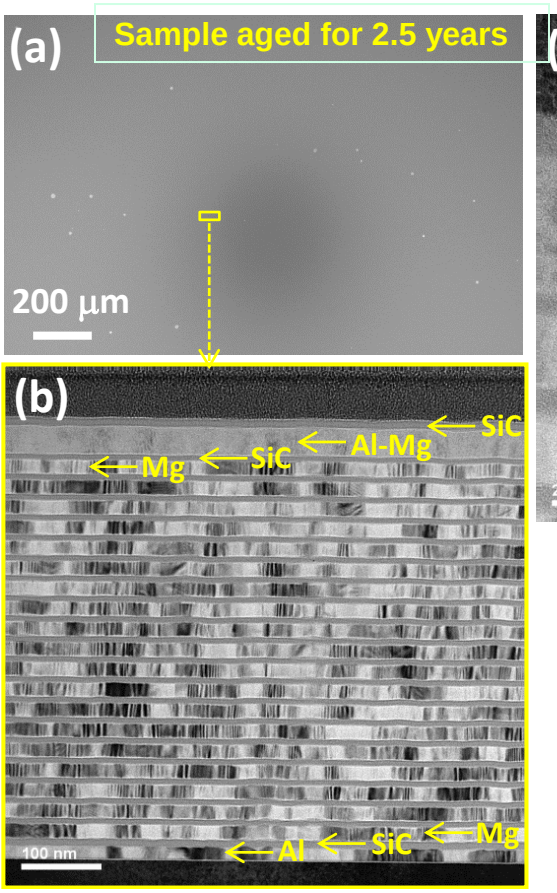


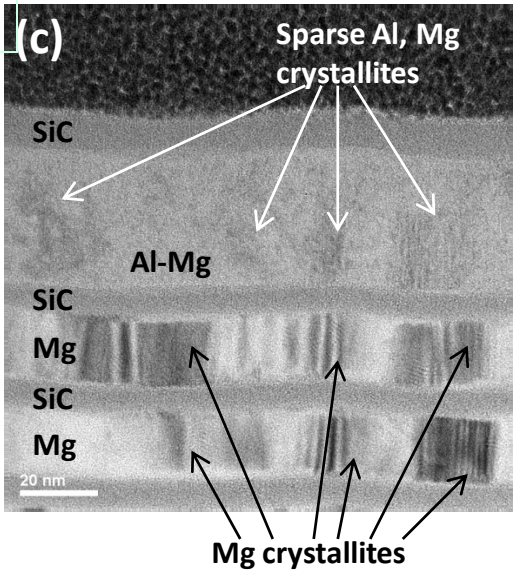
Atmospheric corrosion attacks Mg/SiC from the top surface via localized entry points such as pinholes and defects, inherent in sputtered thin films

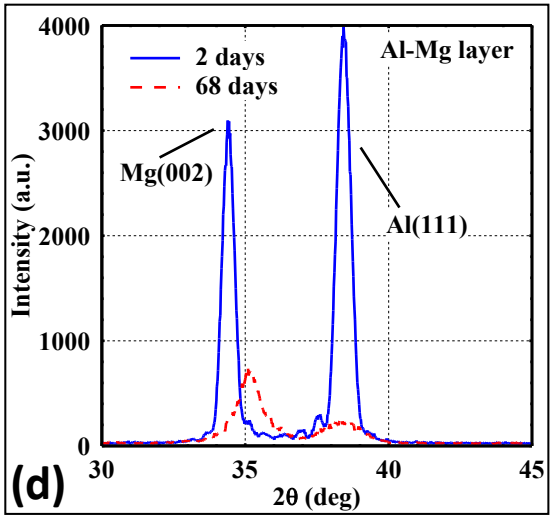
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We have demonstrated Al-Mg corrosion barrier structures for Mg/SiC multilayers

Polycrystalline Al (20 nm) and Mg (19 nm) layers spontaneously intermix to produce partially amorphous Al-Mg layer. Amorphization process takes several weeks to complete.







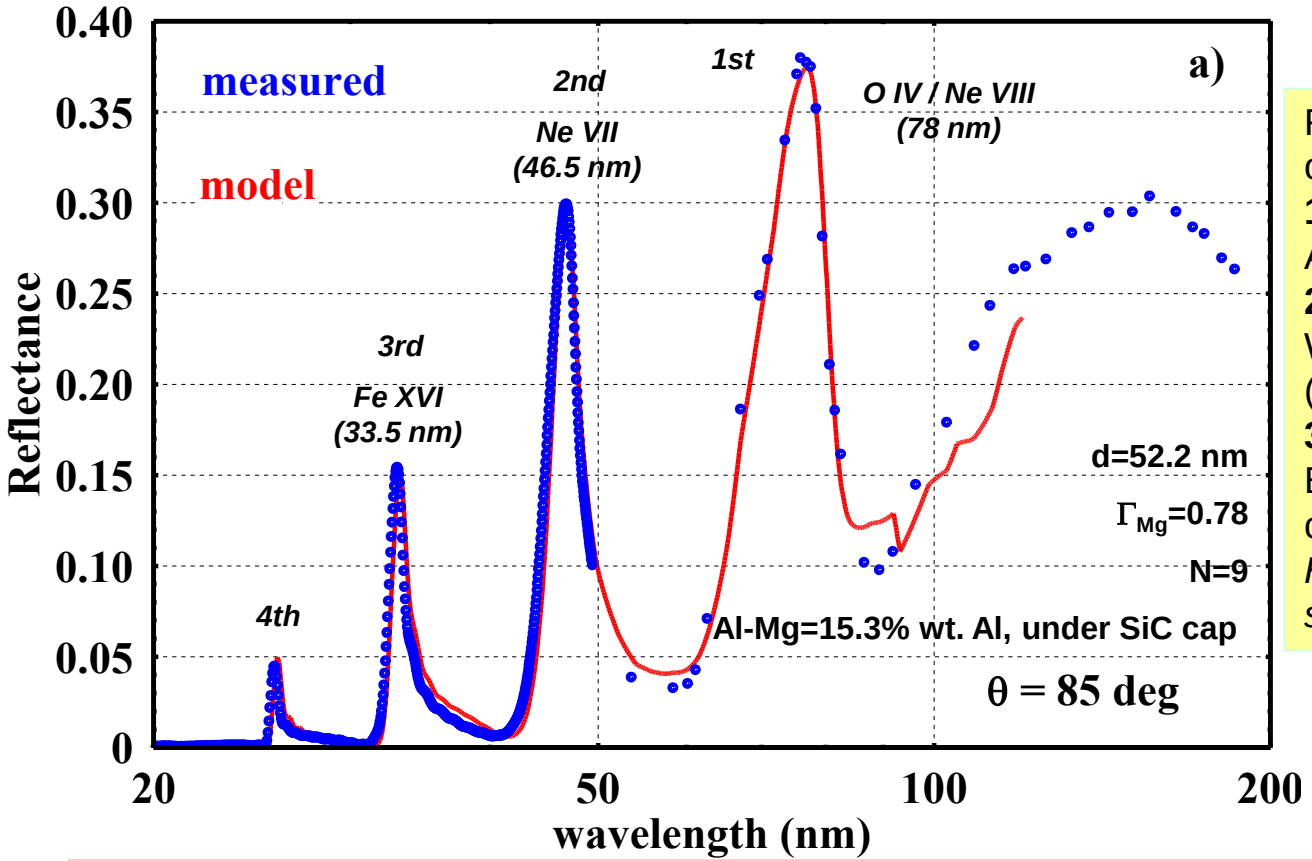
(e)

	Days after deposition	Bragg peak, 2θ (deg)	Lattice spacing (nm)	Crystallite size (nm)
Al(111)	2	38.44	0.23	15.13
”	68	38.41	0.23	4.57
Mg(002)	2	34.41	0.26	16.7
”	68	35.14	0.25	9.15

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Triple-wavelength Mg/SiC multilayers with corrosion barriers can be employed in EUV imaging instruments or spectrometers

Shown below is a periodic (constant d) multilayer. Aperiodic or "stacked" multilayer designs can also be implemented to achieve customized reflective performance. Corrosion barriers can be inserted in specific Mg layers or in each Mg layer



Refractive index values obtained from:

1. Mg: M. Vidal-Dasilva, *et al*, J. Appl. Phys. 108, 063517 (2010).

2. SiC: J. B. Kortright and D. L. Windt, Appl. Opt. 27, 2841-2846 (1988).

3. Al: E. Shiles, *et al,* Phys. Rev. B 22, 1612-1628 (1980), as compiled by E. D. Palik, *Handbook of optical constants of solids* (Academic Press, 1985).

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Summary and future work

- Corrosion has prevented the use of Mg/SiC multilayers in applications requiring good lifetime stability

- We have developed triple-wavelength Mg/SiC multilayers with Al-based barrier layers that dramatically reduce corrosion, while preserving high reflectance and low stress

- We are currently studying in detail the physics of spontaneous intermixing and amorphization of Al and Mg layers

- Corrosion barrier layers should be tested and optimized individually for each multilayer design and environmental conditions

- The aforementioned advances may enable the implementation of corrosion-resistant, high-performance Mg/SiC coatings in applications such as EUV/soft x-ray laser sources and solar physics

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